

Anisotropic Magnetoresistance in Manganites: Model and Experiment

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We present measurements of anisotropic magnetoresistance of $\text{La}_{0.75}\text{Sr}_{0.25}\text{MnO}_3$ films deposited on (001) SrTiO_3 substrates, and develop a model to describe the low temperature AMR in manganites. We measure an AMR of the order of 10^{-3} for the current I parallel to the [100] axis of the crystal and vanishing AMR for $I//[110]$, in agreement with the model predictions.

Introduction - Colossal magnetoresistant manganites have been investigated thoroughly since the discovery of their magnetoresistive properties. [1, 2, 3, 4, 5] Anisotropic magnetoresistance (AMR) of these compounds has also been investigated since it may give rise to application of these materials in electronics.[6, 7, 8, 9, 10] It has been found that the resistivity of polycrystalline ferromagnetic metals and alloys, in the magnetic ordered state, depends on the angle θ between the magnetization M and the electric current I . This dependence has the form[11]

$$\rho(\theta) = \frac{\rho_{\parallel} + 2\rho_{\perp}}{3} + \left(\cos^2\theta - \frac{1}{3}\right)(\rho_{\parallel} - \rho_{\perp}), \quad (1)$$

being ρ_{\parallel} and ρ_{\perp} the resistivity measured with current flowing parallel or perpendicular to the magnetization, respectively. The AMR is defined as

$$\rho_A = \frac{\rho_{\parallel} - \rho_{\perp}}{\frac{1}{3}\rho_{\parallel} + \frac{2}{3}\rho_{\perp}}. \quad (2)$$

The parameters obtained by fitting the experimental results with Eq. (1) are used to calculate ρ_A from Eq. (2), which quantifies the anisotropy of the resistivity.

AMR has been observed in conventional metallic systems and in colossal magnetoresistant materials. However, their sign and temperature dependences are quite different: while in most conventional metals ρ_A is positive[12] and decreases with decreasing magnetization or increasing temperature, in manganites it is an order of magnitude lower, of opposite sign, and its temperature dependence is non monotonic.[6, 7, 8, 9, 10] These differences point to the fact that different mechanisms must be in action in the different materials. The model proposed by Campbell *et al.*[12] based on the scattering of s waves on the d sites of the material has been successful to the understanding of AMR in metallic alloys (describing properly the effects of impurity concentration and temperature dependence) and it has also been mentioned in reference to measurements in manganites. However, it is not appropriate to apply this model to manganites where the carriers (electrons or polarons) move by hopping between the d states of the transition metal. We present here measurements on manganite films and a model that describes the resulting anisotropy as well as its dependence on the direction of the current to the crystalline axes.

Experiment - The ferromagnetic manganite $\text{La}_{0.75}\text{Sr}_{0.25}\text{MnO}_3$ (LSMO) presents a nearly cubic perovskite structure[13] that makes this system appropriate to compare with the model, which is developed for a cubic lattice of Mn ions. The samples were deposited on (001) SrTiO_3 substrates by dc sputtering. The films grow textured following the (001) orientation of the substrate, as confirmed by X-ray diffraction.[14] This substrate, having a lattice constant similar to that of LSMO, induces little distortion on the film compared to the bulk manganite.

We present electrical transport measurements performed on films of different thicknesses, using the longitudinal four-lead configuration with the electrical contacts on the plane of the films. The magnetic field was applied parallel to the plane of the samples with an electromagnet mounted on a rotating platform. All the measurements were carried out with an applied field $H = 10$ kOe, which is strong enough to saturate the magnetization. So the angle θ between the electrical current and the magnetic field is assumed to be the same as the angle between the current I and the magnetization M .

Figure 1 shows the normalized resistivity $[\rho(\theta) - \rho(0)]/\rho(0)$ measured at $T = 88$ K with the current applied parallel to the crystalline direction [100] showing agreement with Eq. (1) and previous AMR measurements.[9, 10] We have not found any systematic dependence on the thickness of the films. Prompted by the model prediction of vanishing AMR we also performed measurements with the current parallel to [110] direction. The resulting AMR is displayed in Fig. 1 by empty circles, where we see negligible dependence of the resistivity with the magnetic field direction. The general formula obtained by Döring[11, 15] for the resistivity of a cubic ferromagnet with the magnetization in the $(\alpha_1, \alpha_2, \alpha_3)$ direction and the current in the $(\beta_1, \beta_2, \beta_3)$ direction is

$$\begin{aligned} \rho = & \rho_0 \left[1 + k_1(\alpha_1^2\beta_1^2 + \alpha_2^2\beta_2^2 + \alpha_3^2\beta_3^2 - \frac{1}{3}) \right. \\ & + 2k_2(\alpha_1\alpha_2\beta_1\beta_2 + \alpha_2\alpha_3\beta_2\beta_3 + \alpha_3\alpha_1\beta_3\beta_1) \\ & + k_3(s - \frac{1}{3}) + k_4(\alpha_1^4\beta_1^2 + \alpha_2^4\beta_2^2 + \alpha_3^4\beta_3^2 + \frac{2}{3}s - \frac{1}{3}) \\ & \left. + 2k_5(\alpha_1\alpha_2\alpha_3^2\beta_1\beta_2 + \alpha_2\alpha_3\alpha_1^2\beta_2\beta_3 + \alpha_3\alpha_1\alpha_2^2\beta_3\beta_1) \right] \end{aligned}$$

where $s = \alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2$. Considering the geometry

studied here, we obtain

$$\frac{\Delta\rho(\theta)}{\rho(0)} \propto \begin{cases} C_1 \cos^2(\theta) + C_3[\cos^4(\theta) - \cos^2(\theta)] & \text{for } I//[100] \\ C_2 \cos^2(\theta) - C_3[\cos^4(\theta) - \cos^2(\theta)] & \text{for } I//[110] \end{cases} \quad (4)$$

where $C_1 = k_1 + k_4$, $C_2 = k_2$ and $C_3 = (k_4 - 3k_3)/3$. The experimental results imply that the constants C_2 and C_3 are almost zero, and the only finite constant is $C_1 = k_1 + k_4$.

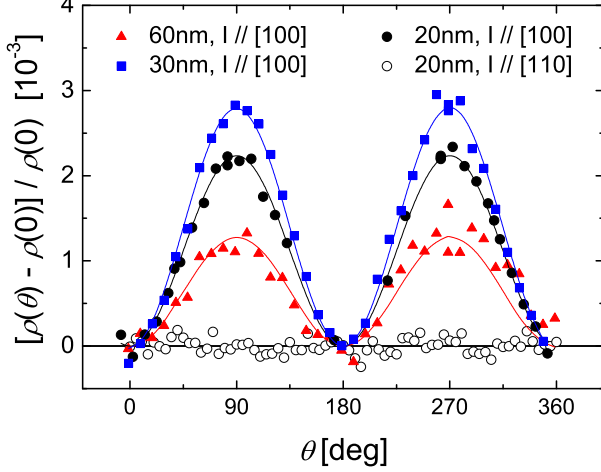


FIG. 1: (Color online) Normalized resistivity measured at $T = 88$ K with the current I applied parallel to the crystalline direction $[100]$ for films of three different thicknesses (20 nm, 30 nm and 60 nm). Hollow circles represent the resistivity measurements with $I//[110]$ for the 20 nm sample. θ is the angle measured between the direction of the electrical contacts and the applied magnetic field ($H = 10$ kOe). A fit with Eq. (1) is also shown (lines).

Model - In $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, the $\text{Mn}^{3+/4+}$ ions form a nearly simple cubic lattice, with oxygen ions located between each pair of Mn neighbors and La/Sr ions at the body center of the cube. The octahedral symmetry around each Mn splits the $3d$ levels into a lower energy t_{2g} triplet and a higher energy e_g doublet. Due to Hund's rule, the three t_{2g} orbitals are all singly occupied with their spins coupled to form a total spin $S = 3/2$. The additional electron on a Mn^{3+} ion occupies the e_g orbitals and it is considered, due again to the strength of the exchange term, to align its spin parallel to the t_{2g} electrons. We model then the e_g electrons by a spinless Hamiltonian on a cubic lattice:

$$H = \sum_{\langle ij \rangle \alpha \beta} t_{ij}^{\alpha\beta} c_{i\alpha}^\dagger c_{j\beta}$$

with $t_{ij}^{\alpha\beta}$ the hopping integrals that depend both on the type of orbitals α, β and on the direction between neighbouring sites i, j . [16] At low temperatures we assume that the localized spins of the t_{2g} electrons are all aligned with the external magnetic field.

When spin-orbit (SO) coupling is included, the degeneracy of the e_g orbitals ($|z\rangle = |3z^2 - r^2\rangle$, $|x\rangle = |x^2 - y^2\rangle$) is lifted. By symmetry, there is no coupling between $e_g \uparrow$ and $e_g \downarrow$ orbitals. Moreover, we take into account only the coupling between the $e_g \uparrow$ and $t_{2g} \uparrow$ orbitals (separated by the crystal field by $\sim 1.5\text{eV}$), and neglect the coupling with the $t_{2g} \downarrow$ orbitals (separated by $\sim 6\text{eV}$ [17]). The character of the two, now not degenerated, orbitals ($|1\rangle, |2\rangle$) depends on the direction of the magnetic field. From second order perturbation theory, the shift and coupling of the two original e_g orbitals for the magnetization in a given direction (θ_B, ϕ_B) are

$$H_1 = g \begin{pmatrix} 3 \sin^2(\theta_B) & \sqrt{3} \sin^2(\theta_B) \cos(2\phi_B) \\ \sqrt{3} \sin^2(\theta_B) \cos(2\phi_B) & \sin^2(\theta_B) + 4 \cos^2(\theta_B) \end{pmatrix} \quad (5)$$

where $g = \lambda^2 / \Delta_{CF}$, being λ the SO coupling constant and Δ_{CF} the crystal field splitting between t_{2g} and e_g orbitals. From this perturbation we obtain the new energy levels ($\varepsilon_{1,2} = g(2 \mp \Delta)$) and the corresponding eigenvectors

$$|1\rangle, |2\rangle = \frac{(a \mp \Delta)}{r_{1,2}} |z\rangle + \frac{b}{r_{1,2}} |x\rangle \quad (6)$$

where

$$\begin{aligned} a &= \sin^2(\theta_B) - 2 \cos^2(\theta_B) \\ b &= \sqrt{3} \sin^2(\theta_B) \cos(2\phi_B) \\ \Delta &= \sqrt{a^2 + b^2} \\ r_{1,2} &= \sqrt{(a \mp \Delta)^2 + b^2}. \end{aligned}$$

Assuming an isotropic relaxation time τ , we calculate the conductivity in a given direction \hat{r} by [18]

$$\sigma_{\hat{r}} = e^2 \tau \int d^3k |v_{\hat{r}}(\vec{k})|^2 \frac{\partial f}{\partial \varepsilon(\vec{k})} \quad (7)$$

with $v_{\hat{r}}(\vec{k}) = \hat{r} \cdot \vec{\nabla} \varepsilon(\vec{k})$ and $f(\varepsilon)$ the Fermi function.

We have taken as the energy reference the hopping t between two $|z\rangle$ orbitals in the \hat{z} direction. Therefore, the only parameter in the model is the constant $g/t = \lambda^2 / (\Delta_{CF} t)$. We take $g/t = 0.001$ to fit the experimental results for the 30 nm film. This value is perfectly consistent with the atomic value of the LS coupling ($\lambda = 0.04\text{eV}$), the crystal field splitting ($\Delta_{CF} = 1.5\text{eV}$) and a hopping of $t = 0.4\text{eV}$. [6]

We first consider the case with the magnetic field rotating in the \hat{x} - \hat{y} plane which corresponds to films with normal $[001]$. We calculate the conductivity in two directions ($[100]$ and $[110]$) and obtain the dependence of the normalized resistivities $[\rho(\theta) - \rho(0)]/\rho(0)$ with the direction of the magnetic field. We show the calculated AMR in Fig. 2, where the angle θ of the magnetic field is measured from the corresponding current direction. Although these calculations were carried out numerically, the result for $I//[100]$ is indistinguishable from a $\cos^2 \theta$ dependence like that of Eq. (1). We can see that, in

agreement with the experimental results shown in Fig. 1, the resistivity for $I // [110]$ does not depend on the direction of the magnetic field. This result can be understood from the $\cos^2(\theta)$ dependence of the resistivity, the cubic symmetry, and the dependence of conductivity with current direction given by Eq. (7). In fact, from the cubic symmetry we have that $\sigma_{[010]}(\theta) = \sigma_{[100]}(\theta - \pi/2)$, and from Eq. (7), we have $\sigma_{[110]}(\theta) = [\sigma_{[100]}(\theta) + \sigma_{[010]}(\theta)]/2$. Then, assuming $\sigma_{[100]}(\theta) = \bar{\sigma} + \Delta\sigma[\cos^2(\theta) - 1/2]$ we obtain $\sigma_{[110]}(\theta) = \bar{\sigma}$, which clearly does not depend on the magnetic field direction θ .

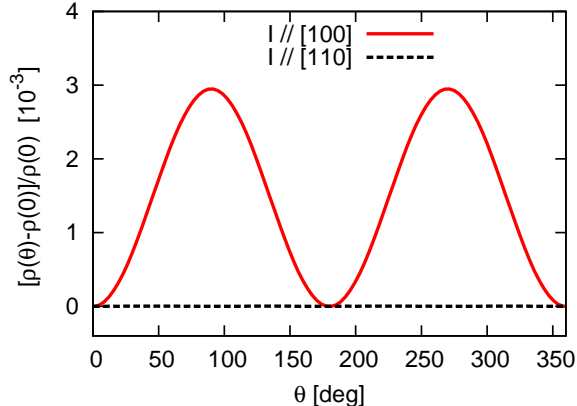


FIG. 2: (Color online) Calculated normalized resistivity in the $[100]$ and the $[110]$ directions, as a function of the magnetic field direction θ measured from the corresponding current direction. The magnetic field rotates in the \hat{x} - \hat{y} plane.

We now consider another case that corresponds to films with normal $[011]$. The magnetic field now rotates in the $[100]$ - $[0\bar{1}1]$ plane and we calculate the conductivity in the two nonequivalent directions $[100]$ and $[0\bar{1}1]$. We show these calculated AMR in Fig. 3, where as before the angle θ of the magnetic field is measured from the current direction. It is difficult to compare this prediction with previous experimental results,[8, 9] since the behaviour of AMR strongly depends on the samples.

In Fig. 4, we show the variation with doping of the AMR defined as $[\rho(\pi/2) - \rho(0)]/\rho(0)$ for the first case (Fig. 2), and the current in the $[100]$ direction. We can see a strong increasing of the AMR with the doping, in particular for $x \lesssim 0.3$. For these calculations we keep all parameters fixed, except for the occupation number $n = 1 - x$. One can expect that both the hopping t and the crystal field splitting Δ_{CF} will increase with the doping. As a consequence the constant g/t , and therefore the AMR, will decrease. Moreover, we have not taken into account neither the effect of the substrate on the film nor the deviation from the cubic symmetry observed in manganites by changing the doping.[13]

Conclusions - We have measured the low temperature anisotropic magnetoresistance of (001) $\text{La}_{0.75}\text{Sr}_{0.25}\text{MnO}_3$

films. We have also formulated a simple model to calculate its angular dependence for different directions of the

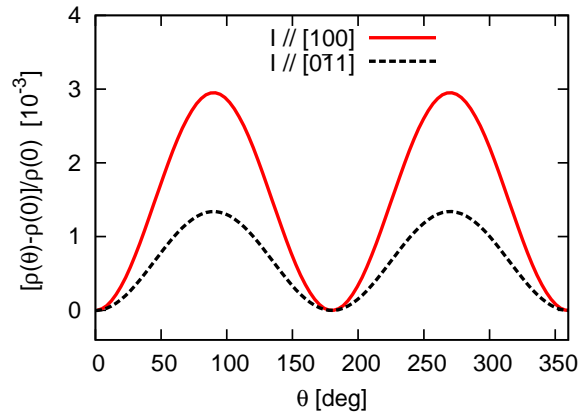


FIG. 3: (Color online) Calculated normalized resistivity in the $[100]$ and the $[0\bar{1}1]$ directions, as a function of the magnetic field direction θ measured from the corresponding current direction. The magnetic field rotates in the $[100]$ - $[0\bar{1}1]$ plane.

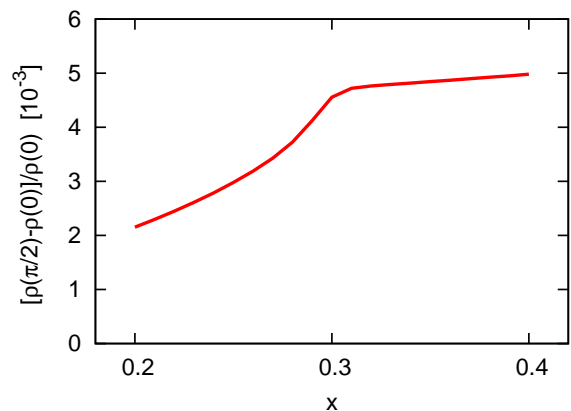


FIG. 4: (Color online) Dependence of the AMR with the doping x .

current to the crystalline axes. The model explains satisfactorily the sign and magnitude of the measured AMR for the current along the $[100]$ axis. Furthermore, it also accounts for the vanishing of the AMR when measured with the current flowing in the $[110]$ direction. More research is necessary to extend the model to describe the dependence of the AMR with doping, temperature, and orientation and matching with substrate.

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